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High Sensitivity Neutron Assay of Grouted Spent Nuclear Fuel Sludge at Hanford

Prepared for the U.S. Department of Energy
Assistant Secretary for Environmental Management
Project Hanford Management Contractor for the
U.S. Department of Energy under Contract DE-AC06-96RL13200

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(Invited)

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INTRODUCTION

The disposal of the North Loadout Pit (NLOP) waste at Hanford will produce 208-liter grouted sludge drums bearing transuranic (TRU) radionuclides and fission products. Discrimination between low level waste (LLW) and TRU waste requires a lower limit of detection (LLD) of less than 100 nCi (3700 Bq) of TRU alpha activity per gram of waste matrix in order to correctly certify the final waste form. Hanford's Waste Receiving and Processing (WRAP) facility operates two identical Imaging Passive Active Neutron (IPAN™) systems which had previously demonstrated this low detection limit capability for debris waste. These two IPAN™ systems were selected as the appropriate technology to assay this challenging waste stream.

In order to meet the required measurement objectives including demonstration of precision and accuracy requirements for the disposal of the waste at the Waste Isolation Pilot Plant (WIPP), the following program of work was undertaken:

a. Design and construction of representative non-radioactive sludge surrogate drums,
b. Calibration of the IPAN™ systems using a novel imaging technique,
c. Testing of the systems using special nuclear materials and high-energy gamma-emitting fission product standards, and
d. Demonstration of the performance requirements under a strict Quality Assurance (QA) program.

The implications of the successful completion of this work have far reaching consequences for the radioassay of remote handled (RH) waste streams. The new method

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offers a viable alternative measurement approach over the "Dose to Curie" method, without the latter method's reliance on operator records and sampling regimes.

The IPAN™ measurement technique comprises active neutron interrogation and passive neutron counting in a single assay chamber. The neutron results are then combined with isotopic information from either acceptable knowledge (AK) or a gamma spectroscopy measurement in order to determine the desired radionuclides. A more detailed description of the principles of active and passive IPAN™ measurements are provided elsewhere [1,2,3]. The passive and active neutron data are processed through a mathematical imaging algorithm. The purpose of the imaging algorithm is to reduce the measurement uncertainty due to source positioning variation within the waste containers. The result is improved sensitivity and regulatory compliant precision and accuracy of results [4].

SLUDGE WASTE CHARACTISTICS

The NLOP sludge is encapsulated in a cement and bentolite matrix within a 208 liter drum. The sludge contains corrosion products from underwater storage of spent nuclear fuel and will therefore contain particles of uranium, fission and activation products and transuranics including plutonium. In order to simulate the sludge, two non-radioactive surrogates were built to the specification given elsewhere [2]. Note that after the specified amount of water was added to the dry sludge, the supernatant was decanted and the precipitate was filtered (but not dried) and mixed with cement. The mixture was poured into a 208 liter drum with a polyethylene liner and allowed to cure. The drum contains 3 polypropylene insert tubes that allow radioactive standards to be loaded with the matrix at various radii and heights [2]. The primary physical difference between the surrogate drums is the amount of their water content. The water content is the most important physical characteristic of the sludge drums with regard to neutron transport.

CALIBRATION TECHNIQUE

Calibration measurements were acquired using the two surrogate sludge drums. A 50 g weapon grade (WG) Pu standard with a ^{240}Pu-effective coincidence mass of 3.03 g was used for passive mode calibration and a 1.0g WG Pu standard with a ^{239}Pu-effective mass of 0.94g was used for active mode calibrations. Measurements were performed with each source positioned at 12 different locations in the drum.

The calibration matrix files were constructed using a radial averaging method. For a given source height, the response factors from each tube were averaged together using weighting factors in order to simulate the effect of source material uniformly distributed across the drum radius. This is done to simulate the expected spatial distribution of source material in homogeneous sludge. As a consequence, the imaging matrix term has 3 (axial) volume elements for each matrix.

DETECTION LIMITS

The LLD in active and passive modes was determined by performing replicate blank measurements on the surrogate sludge drums with and without a 113 mCi (4.18 GBq) ^{137}Cs source in the center of the drum. The active mode LLD has been converted to a nCi/g equivalent minimum detectable concentration (MDC) using various isotopics profiles [5,6,7]. For SNF sludge isotopics, the MDC calculation included a correction for ^{235}U present in these streams. An LLD < 0.061 g ^{239}Pu-effective (< 100nCi/g) in active mode has been successfully demonstrated. In the passive mode, the LLD was approximately 0.04g ^{240}Pu-effective (0.6g WG Pu). The presence of the 137Cs source resulted in only a small increase in the system's active LLD, but had no effect on the passive mode LLD.

CALIBRATION CONFIRMATION

Confirmation measurements were performed for both systems with sludge surrogates covering a wide range of known Pu source loadings. The results are summarized in Table 1. It can be seen from this data that the validity of the calibration is confirmed in active mode and passive mode up to 198 g WG Pu for both sludge matrix surrogates.

Table 1: Sludge Calibration Confirmation Results

WG Pu (g)	Sludge Matrix	Active Mode		Passive Mode	
		%R[a]	*%RSD*[b]	*%R*	*%RSD*
1.9	M1	93.30%	5.70%	N/A	N/A
	M2	87.60%	3.00%	N/A	N/A
9.9	M1	83.40%	1.50%	64.50%	2.80%
	M2	83.20%	3.10%	57.40%	0.90%
198	M1	N/A	N/A	108.30%	2.20%
	M2	N/A	N/A	74.70%	1.50%

[a] Percent Recover, a measure of accuracy
[b] Percent relative standard deviation, a measure of precision

Generally the active mode is most suited for lower ^{239}Pu-effective mass where self-shielding effects are minimal, while the passive mode is best suited for the high ^{240}Pu-effective mass because of improved statistical precision. Note that with homogeneous sludge waste, the effect of self-shielding in active mode is likely to be less severe than for debris measurements.

IMPLICATIONS FOR ASSAY OF REMOTE HANDLED WASTE

An investigation of the impact of high-energy gamma radiation on the system detection limits was performed using a113mCi Cs137 source that produced contact dose rates up to 370 mrem/hr at the drum surface. There was no statistically significant impact on passive mode LLD, and only a relatively small increase in the active mode LLD. These results are particularly encouraging given that WRAP systems were not specifically tailored to deal with intense gamma radiation. The addition of shielding materials and gamma hardened neutron detectors [8] to the IPAN™ design will further improve the applicability of this method to measuring high gamma emitting radioactive wastes, making a good candidate for TRU/LLW sorting of RH waste.

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